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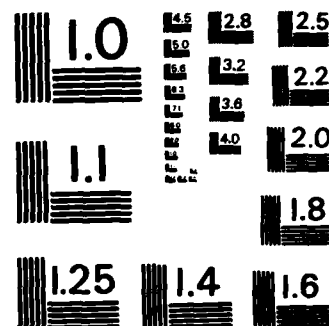
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METAL CLUSTER TOPOLOGY: APPLICATIONS TO GOLD AND PLATINUM CLUSTERS

R.B. King
Department of Chemistry
University of Georgia

ABSTRACT

The graph-theory derived approach for metal cluster bonding is extended to gold and platinum clusters including spherical and toroidal centered gold clusters and stacked triangle platinum clusters; the latter appear to be novel examples of Möbius systems.

INTRODUCTION

In 1977 we published a graph-theoretical interpretation of the bonding topology in delocalized inorganic polyhedral molecules [1]. Our initial treatment [1] focussed on polyhedral boranes, carboranes, and metal clusters. Subsequent work [2] extended these methods to bare metal clusters of post-transition elements such as tin, lead, and bismuth. Further details of our methods are given in a recent book chapter [3]. In general the results of the graph-theory derived methods, insofar as a comparison is possible, are consistent with other approaches to metal cluster bonding by workers such as Mingos [4,5], Stone [6,7], and Teo [8,9,10].

This paper extends our graph-theory derived approach for metal cluster bonding to gold and platinum clusters, which require a variety of new concepts. Mingos [11,12,13] has extended his methods to the treatment of gold clusters but relatively little success has been achieved until now in the understanding of the bonding in platinum clusters. For example, Teo's methods [9] do not give exact electron counts for some of the most common types of platinum carbonyl clusters.



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BACKGROUND

The atoms at the vertices of polyhedral cluster compounds may be light atoms using only s and p orbitals for chemical bonding (e.g., boron or carbon) or heavy atoms using s, p, and d orbitals for chemical bonding (e.g., transition metals or post-transition elements). If these vertex atoms are normal, they use three valence orbitals for intrapolyhedral bonding leaving one or six external orbitals in the case of light or heavy atoms, respectively. The single external orbital of a light vertex atom such as boron or carbon normally bonds to a single monovalent external group (hydrogen, halogen, alkyl, aryl, nitro, cyano, etc.). The six external orbitals of a heavy vertex atom such as a transition metal may be used for a much greater variety of purposes including the following: (1) A single external orbital bonding to a carbonyl, phosphine, or isocyanide ligand; (2) Three external orbitals bonding to a benzene or cyclopentadienyl ring; (3) A single external orbital containing a non-bonding lone electron pair (common for post-transition element vertices).

An important question in polyhedral cluster compounds is whether their chemical bonding is localized along the edges of the polyhedron or delocalized in the surface and volume of the polyhedron. Delocalized bonding occurs when there is a mismatch between the vertex degree of the polyhedron (i.e., number of edges meeting at the vertex) and the number of internal orbitals from the vertex atom. For normal vertex atoms using three internal orbitals there are the following three fundamental cases:

- (A) Planar polygons (all vertices of degree two): Mismatch ($3 \neq 2$) leading to delocalized bonding in planar polygonal aromatic systems such as benzene and cyclopentadienide.
- (B) Simple [14] polyhedra (all vertices of degree three): Match ($3 = 3$) leading to localized bonding such as in polyhedranes (e.g., cubane, dodecahedrane, etc.).
- (C) Deltahedra (all triangular faces) having no tetrahedral chambers (i.e., all vertices of degree four or greater): Mismatch ($3 \neq 4, 5, 6, \dots$) leading to delocalized bonding in three-dimensional aromatic systems such as polyhedral borane anions, carboranes, and many metal clusters. The last case, of course, is the one of greatest interest in the context of this paper.

The three internal orbitals of normal vertex atoms in delocalized polygons or polyhedra can be partitioned into two types: (1) Twin internal orbitals (sp^2 hybrids or p orbitals in a light vertex atom polygon (Case A) or polyhedron (Case C), respectively); (2) Unique internal orbital (p orbital or an sp hybrid in a light vertex atom polygon (Case A) or polyhedron (Case C), respectively). The intrapolyhedral bonding in delocalized deltahedra without tetrahedral chambers and having n vertices requires $2n + 2$ skeletal electrons arising from the following sources:

- (A) Surface bonding ($2n$ skeletal electrons) arising from pairwise overlap (i.e., n K_2 graphs) of the vertex atom twin internal orbitals

in the polyhedral surface to give n bonding and n antibonding orbitals. (B) Core bonding (2 skeletal electrons) arising from n -center overlap (i.e., a single K_n graph) of the vertex atom unique internal orbitals at the polyhedral center to give a single bonding orbital and $n-1$ antibonding orbitals.

Electron-rich delocalized polyhedra having more than $2n + 2$ skeletal electrons form polyhedra having one or more non-triangular faces whereas electron-poor delocalized polyhedra having less than $2n + 2$ skeletal electrons form deltahedra having one or more tetrahedral chambers. A more detailed discussion of bonding models for these systems is given in the previous papers [1,3].

GOLD CLUSTERS

The vertex atoms in the polyhedral clusters treated in our previous papers [1,2,3] use a spherical bonding orbital manifold (sp^3 for light vertex atoms and d^5sp^3 for heavy vertex atoms) having equal extent in all three dimensions leading to the 8-electron (for light atoms) or 18-electron (for heavy atoms) configurations of the next rare gas. However, in some systems containing the late 5d transition and post-transition metals including gold, one or two of the outer p orbitals are raised to antibonding energy levels leading to toroidal (d^5sp^2) or cylindrical (d^5sp) bonding orbital manifolds, respectively. The (d^5sp) toroidal bonding orbital manifold can bond only in the two dimensions of the plane of the ring of the torus leading, for example, to 16-electron square planar complexes of d^8 late transition metals such as Rh(I), Ir(I), Ni(II), Pd(II), Pt(II), and Au(III). Similarly, the (d^5sp) cylindrical bonding orbital manifold can bond only in a single (axial) dimension leading, for example, to 14-electron linear complexes of d^{10} metals such as Pt(0), Ag(I), Au(I), Hg(II), and Tl(III). The p orbitals raised to antibonding energy levels can participate in $d\sigma \rightarrow p\sigma^*$ or $d\pi \rightarrow p\pi^*$ back-bonding with filled d orbitals in adjacent atoms as noted by Dedieu and Hoffman [15] from extended Hückel calculations on Pt(O)-Pt(O) dimers. The raising of one or two outer p orbitals to antibonding levels in heavy late transition metal and post-transition metal complexes has been attributed to relativistic effects [16].

The gold clusters of particular interest [17,18] consist of a center gold atom surrounded by a puckered polygonal belt of peripheral gold atoms generally with one or more additional peripheral gold atoms in distal positions above and/or below the belt. The peripheral gold atoms in such clusters use a 7-orbital d^5sp cylindrical bonding orbital manifold, but their residual two orthogonal antibonding p orbitals can receive electron density from the filled d orbitals of adjacent peripheral gold atoms leading to bonding distances between adjacent peripheral gold atoms. Centered gold clusters can be classified as either spherical or toroidal clusters [19] depending upon whether the center gold atom uses a 9-orbital d^5sp^3 spherical bonding orbital manifold or an 8-orbital d^5sp^2 toroidal bonding orbital manifold, respectively. The topology of the core bonding in the centered gold clusters is generally not that of the

K_n complete graph found in the delocalized deltahedral clusters discussed above but instead corresponds to the topology of the polyhedron formed by the peripheral gold atoms. This apparently is a consequence of the poor lateral overlap of the cylindrical d^{5sp} manifolds of the peripheral gold atoms. Also the volume of the polyhedron of peripheral gold atoms must be large enough to contain the center atom. Thus the icosahedron formed by the twelve peripheral gold atoms in $Au_{13}Cl_2[P(CH_3)_2C_6H_5]_{10}^{3+}$ is regular [20] whereas the cube formed by the eight peripheral gold atoms in $Au_8[P(C_6H_5)_3]_8^+$ is distorted from O_h to D_3 symmetry [21]. This arises from the fact that the internal volume of an Au_{12} icosahedron is large enough to accommodate the center gold atom whereas the internal volume of an Au_8 cube is too small to accommodate the center gold atom. The resulting swelling of the Au_8 cube leads to the observed symmetry reduction.

In the electron counting of centered gold clusters the (neutral) center gold atom is a donor of one skeletal electron, i.e., 11 valence electrons minus the 10 electrons needed to fill its five d orbitals. A toroidal centered gold cluster requires 6 skeletal electrons whereas a spherical centered gold cluster requires 8 skeletal electrons. These numbers are fully consistent with the $12p + 16$ total (skeletal plus external) electron rule for toroidal centered gold clusters and $12p + 18$ total electron rule for spherical centered gold clusters (p is the number of peripheral gold atoms) used by Mingos and co-workers [19]. Such electron counting leads to the general formulas $Au_nL_yX_{n-1-y}^{(y-5)+}$ for toroidal centered gold clusters and $Au_nL_yX_{n-1-y}^{(y-7)+}$ for spherical centered gold clusters where L is a two-electron donor ligand such as phosphine or isocyanide and X is a halide or pseudohalide. Examples of well-characterized toroidal clusters conforming to the $Au_nL_yX_{n-1-y}^{(y-5)+}$ general formula include $Au_8[P(C_6H_5)_3]_7^{2+}$ [22], $Au_8[P(C_6H_5)_3]_8^{3+}$ [23], $Au_9[SCN]_3[P(c-C_6H_{11})_3]_5$ [24], $Au_{10}Cl_3[P(c-C_6H_{11})_2C_6H_5]_6$ [19], and $Au_9[P(C_6H_4OCH_3-p)_3]_8^{3+}$ [25]. Examples of well-characterized spherical clusters conforming to the $Au_nL_yX_{n-1-y}^{(y-7)+}$ general formula include $Au_8[P(C_6H_5)_3]_8^+$ [21], $Au_{11}I_3[P(C_6H_5)_3]_7$ [26], and $Au_{13}Cl_2[P(CH_3)_2C_6H_5]_{10}$ [20].

PLATINUM CARBONYL CLUSTERS

The well characterized large platinum carbonyl clusters fall into the following two categories (Figure 1):

(A) Stacked $Pt_3(CO)_6$ triangles leading to the dianions $Pt_{3k}(CO)_{6k}^{2-}$ ($k = 2, 3, 4, 5$) [27].

(B) Three stacked Pt_5 pentagons (BDB in Figure 1) having a Pt_4 chain (ACCA in Figure 1) inside the stack thereby leading to the $Pt_{19}(CO)_{42}$ cluster [28].

A common feature of both of these types of systems is the stacking of Pt_n polygons leading to a system containing a principal C_n axis on which none of the polygon platinum atoms are located. In the case of the stacked triangle $Pt_{3k}(CO)_{6k}^{2-}$ clusters having such a

C_3 axis, the number of electrons arising from the vertex atoms must be a multiple of 3 or the total number of skeletal electrons must be 2 (mod 3) after allowing for the -2 charge. This requirement alone leads to the $2n + 2$ ($n = 3k$ in this case) skeletal electrons required for delocalized deltahedra but this is not consistent with the observed stacked triangle geometry of the $Pt_{3k}(CO)_{6k}^{2-}$ clusters. Furthermore the height of some of these stacks (i.e., $k = 5$ is known [27]) prevents the unique internal orbitals of all $3k$ vertex platinum atoms from overlapping at the core of the stack in a $3k$ -center bond having the topology of a K_{3k} complete graph.

A bonding model for the stacked triangle $Pt_{3k}(CO)_{6k}^{2-}$ clusters based on the observed geometries and electron counts can incorporate the following ideas:

(A) The $Pt(CO)_2$ vertices are anomalous using four internal orbitals rather than the normal three. They therefore have five external orbitals and are donors of four skeletal electrons each.

(B) The vertices of the interior triangles in the $Pt_{3k}(CO)_{6k}^{2-}$ stack have degree four so that the internal orbitals from these $Pt(CO)_2$ vertices match the corresponding vertex degrees in accord with expectations for edge-localized bonding.

(C) The vertices of the two exterior triangles in the $Pt_{3k}(CO)_{6k}^{2-}$ stack have degree three. After using three internal orbitals of these $Pt(CO)_2$ vertices for edge-localized bonding, there remains one internal orbital from each of the six platinum atoms of the two exterior triangles for further skeletal bonding. Let us call these "extra" internal orbitals on each vertex atom of the exterior triangles the Möbius orbitals.

(D) Edge-localized bonding in each of the $6k - 3$ edges of the $Pt_{3k}(CO)_{6k}^{2-}$ stack requires $12k - 6$ skeletal electrons. Since there are a total of $12k + 2$ skeletal electrons, eight skeletal electrons are left for the two groups of three Möbius orbitals at the top and bottom of the triangle stack. The symmetry of the C_2 axes of the D_{3h} $Pt_{3k}(CO)_{6k}^{2-}$ stacks forces equal allocation of these eight electrons to the top and bottom of the stack. This means that at each end of the $Pt_{3k}(CO)_{6k}^{2-}$ stack there are four electrons for the molecular orbitals formed by the three triangularly situated Möbius orbitals. This electron count suggests that at the top and bottom triangles of the $Pt_{3k}(CO)_{6k}^{2-}$ stacks, there is $4m$ electron (m is an integer, namely one in this case) Möbius overlap involving a twisted ring (Möbius strip) of the three relevant orbitals rather than $4m + 2$ electron untwisted Hückel overlap found in planar aromatic hydrocarbons such as benzene [29]. If the Möbius orbitals are d orbitals, then twisted Möbius overlap is possible for an odd number of metal atoms (e.g., a triangle or pentagon, but not a quadrilateral) since d orbitals change phase (i.e., "twist") at each metal cluster.

This bonding model for the $Pt_{3k}(CO)_{6k}^{2-}$ stacked triangle clusters suggests edge-localized bonding along the $6k - 3$ edges of the stack coupled with delocalized Möbius triangles at both the top and bottom

of the stack. Thus the edge-localized bonding in the $\text{Pt}_{3k}(\text{CO})_{6k}^{2-}$ clusters corresponds to the edge-localized carbon-carbon σ -bonding in benzene whereas the Möbius bonding at the top and the bottom of the $\text{Pt}_{3k}(\text{CO})_{6k}^{2-}$ stack corresponds to the Hückel π -bonding in benzene.

The structure of the threaded tubular platinum carbonyl cluster $\text{Pt}_{19}(\text{CO})_{22}^{4-}$ (Figure 1) can be built as follows:

(1) Three Pt_5 pentagons (BDB in Figure 1) are stacked on top of each other forming two pentagonal prismatic chambers sharing a pentagonal face.

(2) A linear Pt_4 chain (ACCA in Figure 1) is placed on the C_5 axis of the stacked pentagons so that the two end members of the Pt_4 chain are the apices of pentagonal pyramids at the top and bottom of the pentagonal stack and the two central members of the Pt_4 chain are located in the centers of the two pentagonal prismatic chambers noted above.

In this structure of $\text{Pt}_{19}(\text{CO})_{22}^{4-}$ the internal orbitals from the platinum atoms are used as follows:

(A) End platinum atoms of the Pt_4 chain (two platinum atoms): Three internal orbitals are used for a delocalized pentagonal pyramid and the fourth internal orbital is used for a localized bond to the nearest interstitial platinum atom also in the Pt_4 chain.

(B) Platinum atoms of the top and bottom Pt_5 pentagons (ten platinum atoms): Three internal orbitals are used for a delocalized pentagonal pyramid and the fourth internal orbital is used for a localized bond to the nearest platinum atom of the middle Pt_5 pentagon.

(C) Interstitial platinum atoms (the two center platinum atoms of the Pt_4 chain): All nine platinum valence orbitals are internal orbitals so that all of the ten valence electrons of each interstitial platinum atom become skeletal electrons.

(D) Platinum atoms of the middle Pt_5 pentagon (five platinum atoms): All four internal orbitals are used for edge-localized bonds to neighboring platinum atoms.

This allocation of platinum internal orbitals leads to the following electron counting scheme for $\text{Pt}_{19}(\text{CO})_{22}^{4-}$:

Source of skeletal electrons:

17 PtCO vertices using 4 internal orbitals: $(17)(2) =$	34 electrons
5 "extra" CO groups: $(5)(2) =$	10 electrons
2 interstitial platinum atoms: $(2)(10) =$	20 electrons
-4 negative charge on anion	<u>4 electrons</u>
Total available skeletal electrons	68 electrons

Use of skeletal electrons:

Edge-localized bonding in the Pt_{15} tube: 25 edges =	50 electrons
Edge-localized bonding in the Pt_4 chain: 3 edges =	6 electrons
Incremental electrons for the two delocalized pentagonal pyramidal chambers: $(2)[(2)(6)+4-10] =$	<u>12 electrons</u>
Total skeletal electrons required	68 electrons

This indicates that the anion $\text{Pt}_{19}(\text{CO})_{22}^{4-}$ has exactly the number of electrons required for the above bonding model. Furthermore, the existence of a homologous series of threaded tubular clusters is predicted having the general formula $\text{Pt}_{6n+1}(\text{CO})_{6n+4}^{4-}$.

SUMMARY

This paper shows how our graph-theory derived approach for metal cluster bonding can be extended to gold and platinum clusters exhibiting new structural features. For the treatment of these systems important ideas new to this theory need to be introduced, notably non-spherical (i.e., toroidal and cylindrical) bonding manifolds for the gold clusters and Möbius bonding for the stacked triangle $\text{Pt}_{3k}(\text{CO})_{6k}^{2-}$ clusters.

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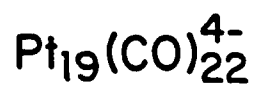
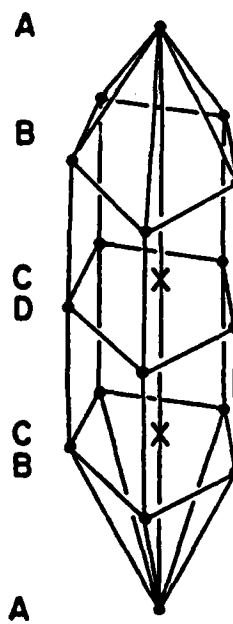
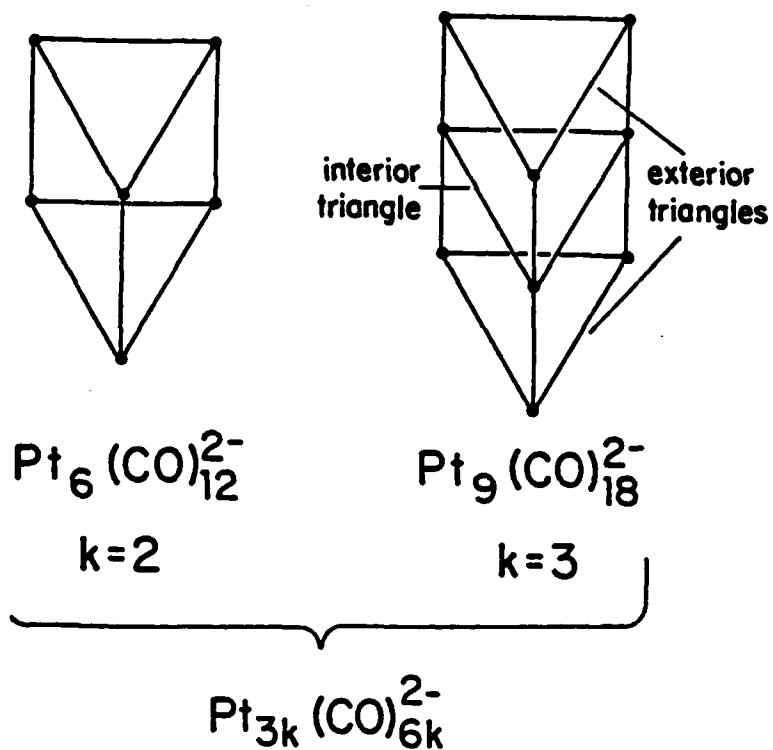


Figure 1: Schematic diagrams of the platinum carbonyl clusters discussed in this paper.

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